Slow Relaxation Process in Ising like Heisenberg Kagome Antiferromagnets due to Macroscopic Degeneracy in the Ordered State

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Abstract. We study relaxation phenomena in the macroscopically ordered state in the Ising like Heisenberg kagome antiferromagnets. In recent experiments, slow relaxation phenomena have been observed in kagome compounds. We introduce the "*weathervane loop*" in order to characterize the ordered state and study the microscopic mechanism of the slow relaxation from a view point of the dynamics of the weathervane loop configuration.

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1. Introduction

Due to the frustration among the magnetic interactions, antiferromagnetic spin systems on the triangular, kagome, or pyrochlore, *etc.* show very interesting properties of magnetic ordering. In particular, in the kagome lattice and the pyrochlore lattice, which are the so-called corner-sharing lattice where neighboring local triangles share a spin but not a bond, frustration causes macroscopic degenerate states in the ground state even for the continuous spin systems, *e.g.* XY, and Heisenberg systems. Thus, there is no phase transition at finite temperatures in the kagome antiferromagnets with Ising, XY and Heisenberg spin systems. However, Kuroda and Miyashita pointed out that the kagome aniferromagnet has a magnetic phase transition with the universality class of the ferromagnetic Ising model [1], when the system has finite easy-axis anisotropy, namely the Ising like Heisenberg interaction. There the ordered state is still macroscopically degenerated, and was called exotic ferromagnetic phase. Recently slow relaxation phenomena were found in the kagome and pyrochlore lattice [2],[3]. In this paper we consider the origin of the slow relaxation from the view point of structural change of the macroscopically degenerate ordered states of the Ising like Heisenberg kagome antiferromagnet.

There are various candidate materials for the antiferromagnetic kagome spin system, for example, $Rb_2M_3S_4$ [2]-[7] (M is a magnetic ion such as Ni, Co, Mn), $AM_3(OH)_6(SO_4)_2$, (A^+ is a cation such as K^+, Rb^+, NH_4^+, Tl^+, Ag^+ or Na^+, and M^{3+} is a magnetic ion such as Fe^{3+}, or Cr^{3+}) [8]-[12], $NaFe_3(SeO_4)_2(OH)_6$ [13], $SrCr_{9x}Ga_{12-9x}O_{19}$ (SCGO) [3], and ^{3}He on the sheet of graphite [14]. Maegawa *et.al.* discovered the two cusps of the temperature dependence of the susceptibility in $NH_4Fe_3(OH)_6(SO_4)_2$ [8]. They concluded that the successive phase transition may be caused by a small Ising-like anisotropy in the Heisenberg kagome antiferromagnet. This result is supporting Kuroda and Miyashita's scenario. In recent experimental studies, the antiferromagnetic kagome compounds shows the slow relaxation of magnetization and magnetic susceptibility. Usually, the slow dynamics is caused by random interaction of the systems. However, the slow relaxation appears in non-random spin systems such as $SrCr_{9x}Ga_{12-9x}O_{19}$ (SCGO), $AM_3(OH)_6(SO_4)_2$ which are corner sharing structure.

In the present paper, we study relaxation phenomena in kagome antiferro magnetic systems in the macroscopically degenerate ordered state. In order to characterize the degenerate state we introduce "weathervane loop", and investigate the microscopic mechanism of the slow relaxation in kagome antiferromagnetic systems by considering relaxation of configuration of the weathervane loop structure, which is much slower than the relaxation of the total magnetization which is the order parameter of the model.

In Section 2, we review the phase transition and features of the Ising like Heisenberg antiferromagnetic kagome systems. In Section 3, we consider relaxation processes of the number of 'weathervane loop'. In Section 4, we conclude our research.

2. Model

We consider the antiferromagnetic Ising like Heisenberg kagome antiferromagnetic system,

$$\mathcal{H} = J\left(\sum_{\langle i,j\rangle} S_i^x S_j^x + S_i^y S_j^y + A S_i^z S_j^z\right), \quad J > 0 \text{ and } A > 1, \tag{1}$$

where $\langle i,j\rangle$ denotes nearest neighbor in kagome lattice and A denotes Ising like anisotropy. Kuroda and Miyashita have shown that the system has a phase transition which belongs to the universality class of two dimensional ferromagnetic Ising spin system [1]. The kagome system consists of triangle unit that shares one corner. The ground state of Ising like Heisenberg triangle unit is given by $S_\alpha = (0,0,1)$, $S_\beta = (s,0,-c)$, $S_\gamma = (-s,0,-c)$, where $c = \frac{A}{A+1}$ and $s = \sqrt{1-c^2}$. The freedom of the rotation 2π in the xy plane remains in S_β and S_γ.

It should be noted that this system has nonzero magnetization in the ground state, the value of which is $1-2c$ in each triangle unit. The magnetization of the ground state per spin is $\frac{M_0}{N} = m_0 = \frac{1}{3}\times(1-2c)$, where M_0 and N denote the total magnetization of the ground state and the number of spins, respectively. It is important to note that no sublattice long range order in this system in spite of the existence of the magnetic phase transition.

In order to study the equilibrium properties, we use the heat bath method of Monte Carlo simulations with $100,000$ Monte Carlo Steps(MCS) for initial relaxation and $100,000$ MCS for collecting data. For the region near the critical point, we perform $200,000$ MCS for initial relaxation and $500,000$ MCS for measurement. Figures 1 show temperature dependence of (a) the magnetization and (b) the specific heat for $A = 3$.

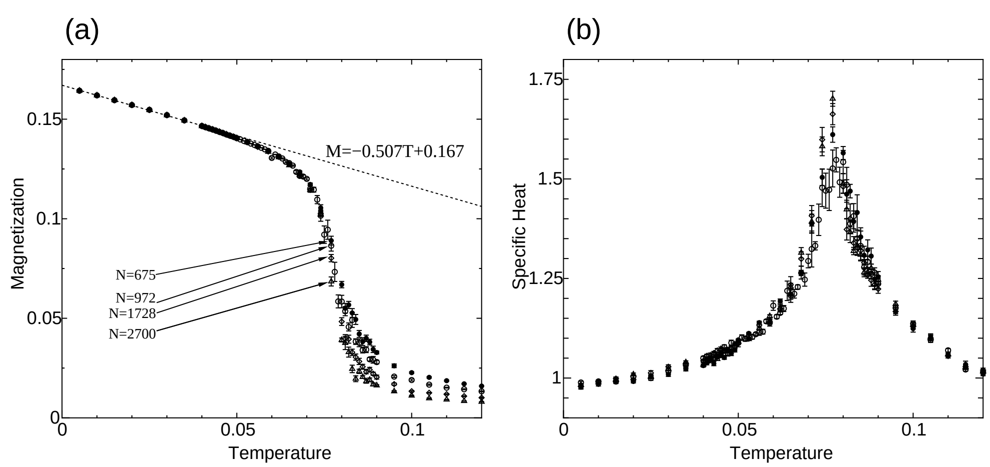

Figure 1. Temperature dependence of (a) the magnetization and (b) the specific heat for $A = 3$. (● for $N = 675$, ○ for $N = 972$, ◇for $N = 1728$, and △for $N = 2700$)

When $T \to 0$, the magnetization approaches the ground state $m_0 = -\frac{1}{6}$. At $T \simeq 0.078$, the magnetization changes suddenly and the specific heat diverges, which

indicates the second order phase transition. This is the phase transition which belongs to the two dimensional Ising ferromagnetic universality class [1].

3. Weathervane Loop and Defects

Because the antiferromagnetic kagome lattice system has large number of degenerated states, it is important to consider the entropy of the spin configuration. If there is the easy-axis type anisotropy (*i.e.* $A > 1$), one spin (S_α) is parallel to the z-axis and two spins (S_β,S_γ) direct opposite direction in each triangle cluster in the ground state. The spins $\{S_\beta\}$ and $\{S_\gamma\}$ have the freedom of the 2π rotating in the xy plane. If we connect $\{S_\beta\}$ and $\{S_\gamma\}$ in the lattice, we find a closed loop as shown in Fig. 2. We call the line *weathervane loop.*

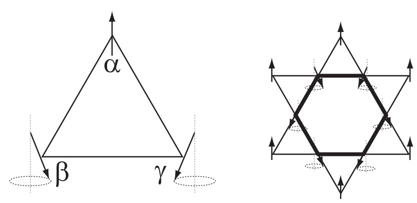

Figure 2. The spin configuration of the ground state in a triangle cluster and an example of the weathervane loop (bold line).

In the ground state, there are macroscopic degenerate configurations of the weathervane loop in Fig. 3(a),(b), and (c). Figs. 3(a) and (b) show the ground state which are called $q = 0$ and $\sqrt{3} \times \sqrt{3}$ structure, respectively. On the other hand, in Fig. 3(c), we show one which obtained by quenching the system from high temperature. This is one of the degenerate configuration and we call it "random structure". Let us consider the degeneracy of the ground state. Each loop has the rotation degree of freedom 2π. We denote the number of the degeneracy of the ground state $(2\pi)^{n_{\rm loop}}$, where $n_{\rm loop}$ denotes the number of weathervane loop. Here we take the degeneracy of single angle to be 2π. In the spin configuration of the $\sqrt{3} \times \sqrt{3}$ structure, $n_{\rm loop}$ takes the maximum value $n_{\rm max} = N/9$.

Next, we consider the time evolution of the number of the weathervane loop and the magnetization. Here we study the system of $A = 3$ and $N = 2700$. In Fig. 4(a), we show time evolution of the magnetization at $T = 0.07$ starting from the initial three types of spin states, *i.e.* random, $q = 0$, and $\sqrt{3} \times \sqrt{3}$. Here the data are obtained as an average over 36 independent runs. The errorvars are of order 0.05 which are omitted. Though the magnetization relaxes to the equibrium value in a time, $\tau_{\rm mag}^{(T=0.07)} \sim 10^3$ MCS, it takes $\tau_{\rm loop} \sim 10^5$ MCS for $n_{\rm loop}$ to relax (Fig. 4(c)). In the inset the initial part is magnified, we find the different relaxation processes in the three cases. In the case of random pattern, we find a simple exponential type relaxation. From $\sqrt{3} \times \sqrt{3}$ structure, the $n_{\rm loop}$ reduces from the maximum value to the equilibrium value monotonically. On the other hand, from $q = 0$ structure, $n_{\rm loop}$ shows an non-monotonic behavior.

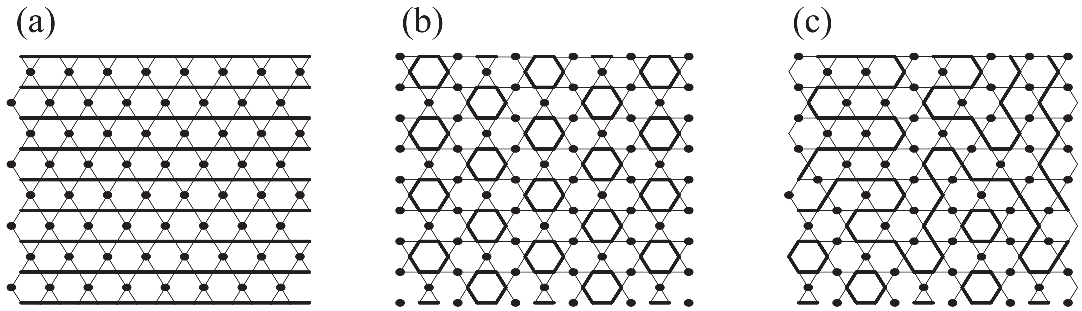

Figure 3. The typical example of the ground state of the anisotropic Heisenberg kagome system. (a)$q = 0$ state, (b)$\sqrt{3} \times \sqrt{3}$ state, and (c)the random ground state. The circles and thick lines denote the S_α and the weathervane loop, respectively.

At $T = 0.05$, the relaxation time of the magnetization is $\tau_{\rm mag}^{(T=0.05)} \sim 10^3$ MCS same as the case of $T = 0.07$(Fig. 4(b)). In this case, it should be noted that the relaxation time of the weathervane loop is more than 10^7 MCS(Fig. 4(d)). This slow relaxation is attributed to the cost that is necessary for the re-arrangement of the weathervane loop. This cost is large comparing to the temperature. The difficulty of the re-arrangement of the weathervane loop is the origin of the slow relaxation in easy-axis type anisotropic Heisenberg kagome antiferromagnets.

4. Conclusion

We consider the origin of the slow dynamics in anisotropic Heisenberg kagome antiferromagnets. In this study, the main stress falls on the difficulty of the re-arrangement of the weathervane loop, which is the origin of the slow relaxation in this system. In this paper, we consider two dimensional kagome antiferromagnetic classical spin systems with Ising like anisotropic interaction. Below the critical temperature, the relaxation of the $n_{\rm loop}$ is slower than of the magnetization which is the order parameter of this model. We expect to realize our scenario in the easy-axis type anisotropic Heisenberg antiferromagnets in the ferromagnetic ordered state. When the inter-plane interaction exists, we have to study the present mechanism in three dimensions. There the weathervane loop becomes weathervane plane and the recombination will have more significant effect on the relaxation, which will be reported elsewhere [15].

Acknowledgments

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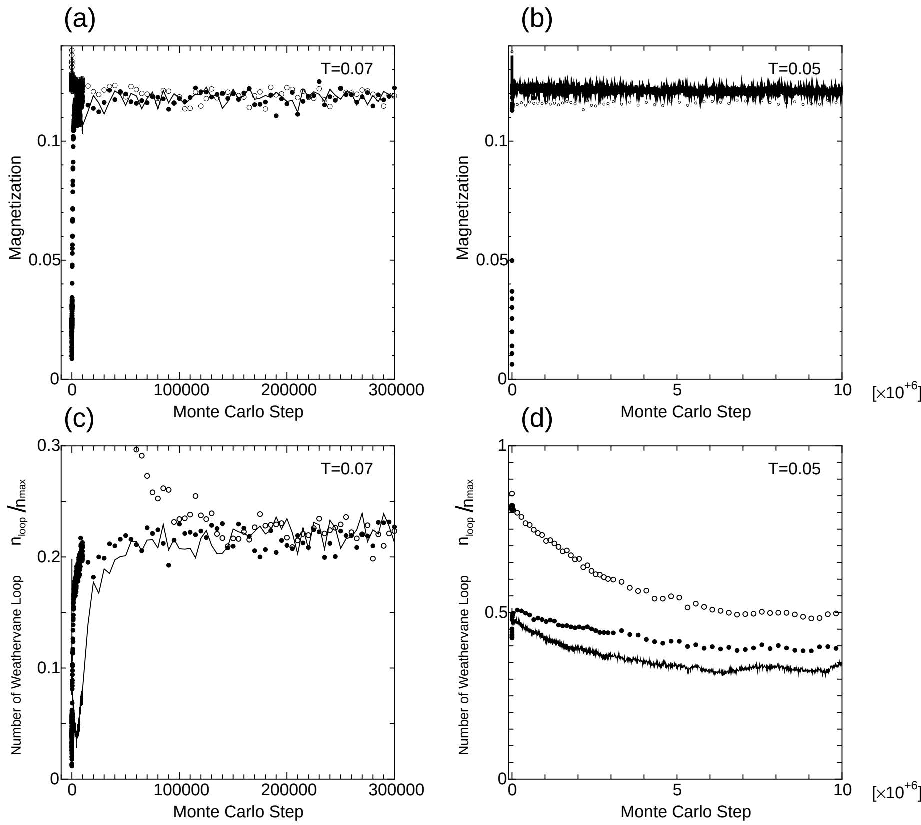

Figure 4. Time evolution of the magnetization (a) at $T = 0.07$ and (b) at $T = 0.05$, and n_{loop} (c) at $T = 0.07$ and (d) at $T = 0.05$. Symbols denote the initial conditions as (○ for $\sqrt{3} \times \sqrt{3}$, a solid line for $q = 0$, and ● for the random).

References

[1] Kuroda A, Miyashita S 1995 *J. Phys. Soc. Japan* **64** 4509.
[2] Wills A S, Dupuis V, Vincent E, Hammann J, Calemczuk R 2000 *Phys. Rev. B* **62** R9264.
[3] Ladieu F, Bert F, Dupuis V, Vincent E, Hammann J 2004 *J. Phys.: Condens. Matter* **16** 5735.
[4] Kato T, Hondou K, Iio K 1998 *J. Magn. Magn. Mater.* **177-181** 591.
[5] Hondou K, Fujiwara Y, Kato T, Iio K, Saiki A, Usuda M, Hamada N 2002 *Journal of Alloys and Compounds* **333** 274.
[6] Hasegawa T, Inui M, Hondou K, Fujiwara Y, Kato T, Iio K 2004 *Journal of Alloys and Compounds* **364** 199.
[7] Kuki R, Fujiwara Y, Ono T, Iio K, Tachibana M, Atake T, Nakai Y 2005 *Prog. Theor. Phys. Suppl.* **159** 61.
[8] Maegawa S, Nishiyama M, Tanaka N, Oyamada A, Tanano M 1996 *J. Phys. Soc. Japan* **65** 2776.
[9] Morimoto T, Nishiyama M, Maegawa S, Oka Y 2003 *J. Phys. Soc. Japan* **72** 2085.
[10] Wills A S, Harrison A, Ritter C, Smith R I 2000 *Phys. Rev. B* **61** 6156.
[11] Nishiyama M, Maegawa S, Inami T, Oka Y 2003 *Phys. Rev. B* **67** 224435.
[12] Inami T, Nishiyama M, Maegawa S, Oka Y 2000 *Phys. Rev. B* **61** 12181.
[13] N. Oba N, Michioka C, Kato M, Yoshimura K, Mibu Ko 2005 *J. Phys. Chem. Sol.***66** 1438.
[14] Graywall D S, Busch P A 1989 *Phys. Rev. Lett.* **62-16** 1868.
[15] Tanaka S, Miyashita S *in preparation.*